RUHR-UNIVERSITÄT BOCHUM
FAKULTÄT FÜR PHYSIK UND ASTRONOMIE

50 Jahre RUB

Today's Polarized Solid Targets in Borghini's Footsteps

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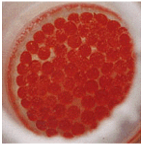
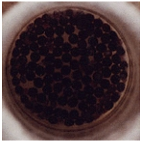
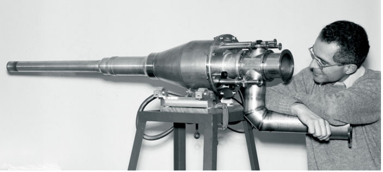
Beijing 2014-10-23







Overview

- Michel Borghini – 'at ease with theory and being in the laboratory'
  - some personal reflections
- Present polarized targets for particle physics experiments using
  - alcohol materials
  - frozen spin technology
- Next challenge
- Spin off – Polarized Target for medical applications

Michel Borghini

some personal reflections

1978 My first contact with Michel at CERN, just before he left the CERN's polarized target group
'Polarized target was an adventure of my youth'

1980 My second contact at the Spin Symposium in Lausanne

Contributions in parallel sessions:
a) Polarized Targets
CHAIRMAN: M. Borghini, CERN

EXPERIENCE WITH NH_3 AS TARGET MATERIAL FOR POLARIZED PROTON TARGETS AT THE BONN 2.5 GEV ELECTRON SYNCHROTRON

U. Härtel, O. Kaul, W. Meyer, K. Rennings, E. Schilling
Physikalisches Institut der Universität Bonn, Germany

Michel Borghini

some personal reflections

1979 – Several young people followed Michel's footsteps

– Ammonia as polarized target material was at the horizon

T. Niinikoski and J.-M. Rieubland Phys.Lett.72A(1979)14
W.Meyer et al, Proc.of.the Int.Symp. High Energy Phys.Pol.Beans and Targets, Basel, Birkhäuser, 1981 p.447 and p.451
D.G. Crabb et al., Proc.of High Energy Spin Phys., Brookhaven 1982, AIP Proc.No.95,1982,p.488

⟶ Talk of D.G. Crabb

Dynamic Polarized Solid Target

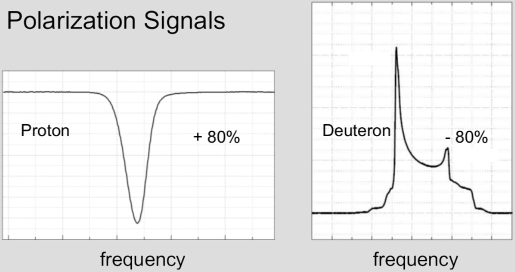
Method:

Production of a high polarization degree in a suitable material **with a high content** on **polarizable nucleons** and **'free' electrons (radicals)** by means of

- **high magnetic field**
- **extrem low temperature**
- **microwave irradiation** → (dynamic nuclear polarization (DNP))

Polarization detection by **Nuclear magnetic Resonance (NMR) technique**

Polarization Signals



In addition: Secondary qualities

- radiation hardness of the polarization
- easy handling – fast target material exchange or polarization refreshment

Situation of polarized target materials:

- 1958 A. Abragam invented the SOLID EFFECT for nonmetal substances

  ↳ triggerd by the Overhauser Effect (1953)

  ↳ Polarization of nuclei in metals

SOLID EFFECT $\longrightarrow$ Dynamic Nuclear Polarization (DNP)

- 1962 First polarized target for particle physics experiments in SACLAY
  - 20MeV pol. protons on pol. protons in LMN-Nd^{3+} crystals
  - $\mathbf{P_t} = 70\,\%$; **f**=3 %; poor radiation damage resistance of the polarization

  ↳ ratio of pol. to unpol. nuclei

1. Reminder: $\boxed{FOM_{target} \sim \mathbf{f}^2}$

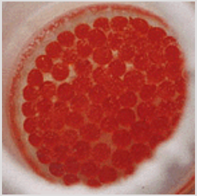
Michel Borghini:
'at ease with theory...' (1)

- 1965: Michel Borghini moved from Saclay to CERN

  'better' pol. target materials was a priority of research

  (Borghini – CERN Yellow Report 66-3,1966, 'Choice of substances for pol. proton targets')
- 1968: Breakthrough with a Butanol+5 % H_2O + Prophyrexide mixture (f=13.5%)

  (S. Mango ,Ö. Runolfsson, M.Borghini, NIM72(1969)456)



Target material for better cooling
in form of beads (1–2mm)

Michel Borghini:
'at ease with theory...' (2)

- 1968: M.B. new phenomenological model of DNP
  'Spin temperature model' $\longrightarrow$ '**Borghini Model**'

  Up to that time:
  - Provotorov theory correctly describes the behaviour of dipolar coupled spin systems under saturation
  - With the exception of the solid state rate equations so far the considerations were restricted to the 'so called' High Temperature Approximation
  - This assumption is far from being valid under usual conditions of a DNP experiment
  - Within this framework an expression for the spin temperature can be derived (degree of saturation; **width of the resonance line**; time constants)

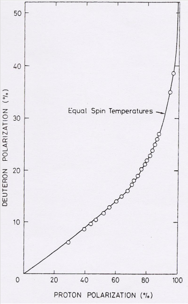
  - **M.B. also found a way to experimentally test the predictions in the low temperature regime**

Michel Borghini:

'.. and being in the laboratory' (1)

Samples: Partially and fully deuterated alcohols and diols

↳ proton pol. compared to the deuteron or ^{13}C polarization



Substance (chemical formula)	Paramagnetic centres (concentration in spins/cm³)	Maximum polarization at 25 kG				Maximum proton polarization at 50 kG below 1.5 K	References
		∿ 1 K	≲ 0.5 K				
		P(H)	P(H)	P(D)	P(C)		
Ethanediol ($C_2H_6O_2$)	Cr^V-complexes $5 \times 10^{19} - 10^{20}$	50	80-97	40	48	80	25-27, 61 95-97
1,2-propanediol ($C_3H_8O_2$)	Cr^V-complexes $5 \times 10^{19} - 2 \times 10^{20}$	50	80-98	44	52	90	28, 29, 32, 98, 99
Hexanediol Pinacone ($C_6H_{14}O_2$)	Cr^V-complexes $10^{19} - 10^{20}$	40	60-80	-	-	-	78, 92, 94
1-butanol ($C_4H_{10}O$)	Porphyrexide 3×10^{19}	40	70-85	25	21	80	100-108
Ammonia (NH_3)	Cr^V-complexes $\sim 10^{19}$	40	60-70	-	-	-	108, 109

⟶ Polarized Solid Targets came into fashion in every particle physics laboratory in the 1970s and 1980s

Michel Borghini:

'.. and being in the laboratory' (2)

Target temperature T and magnetic field B: $\mathbf{P_t} \sim \frac{\mu \mathbf{B}}{\mathbf{kT}}$

2. Reminder: $FOM_{target} \sim \mathbf{P_t}^2$

years	T[K]	B[T]
1960s	1.0	1.8
1970s	0.5	2.5
>1974	<0.1	2.5

↳ CERN: 1. Frozen Spin Target

‘frozen spin polarized target’

‘a real one’ – Niinikoski (CERN) 1974

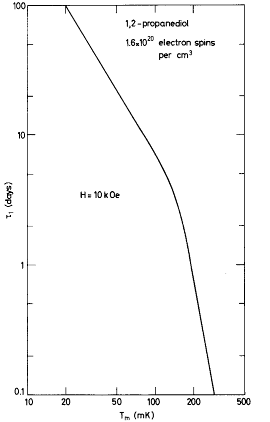
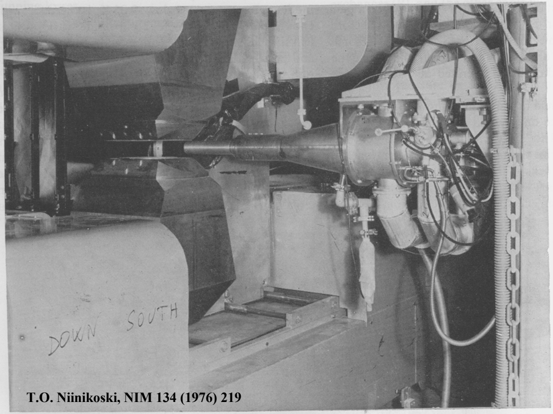


Fig. 7 The proton spin lattice relaxation time interpolated from Ref. 8. T.O. Niinikoski, NIM 134 (1976) 219



T.O. Niinikoski, NIM 134 (1976) 219

Fig. 4. Photograph of the installation. The target is in position D, retracted but on the beam line. The particle beam enters from right, traverses the cryostat and disappears into the magnet spectrometer.

B_{fsp} = 1 T (spectrometer field)

$\tau_{(propandiol)}$ ~ 40 days @ 50 mK

nealy 4π angular acceptance

State-of-the-Art DNP Solid Targets

Two schemes

A Continuous microwave driven DNP targets

- **a** Brookhaven/SLAC-type: 1K; 5T → see D.G.Crabb

  by pure ^{4}He-pumping (superfluid) → high cooling power

- **b** CERN-type: < 0.1K; 2.5T

B Frozen spin targets

- **1** DNP-mode: 0.2-0.3K; 2.5-6T (target material dependent)

  ↓

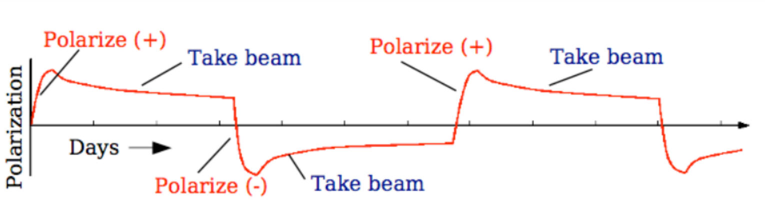
- **2** Frozen spin-mode: < 70mK; 0.4-1.0T

Frozen Spin Target for 4π-Particle Detection (Bonn Type)

Operation is more complicated:

1) Polarize target (DNP) at 2.5/5.0 Tesla and < 0.5 Kelvin;
2) ‘Freeze’ the spins at a very low temperature < 70mk;
3) Use a smaller magnet **(inside the refrigerator)** to hold the polarization (0.5 Tesla) with beam on target;
4) Repeat 1-3 as needed

(Polarization decay time depends on material, field & temperature)



Advantage

Smaller magnet obscures fewer scattered particles

Disadvantage

Very low temperature required for ‘freezing’ the spins means lower beam intensity (~10^7 particles/s)

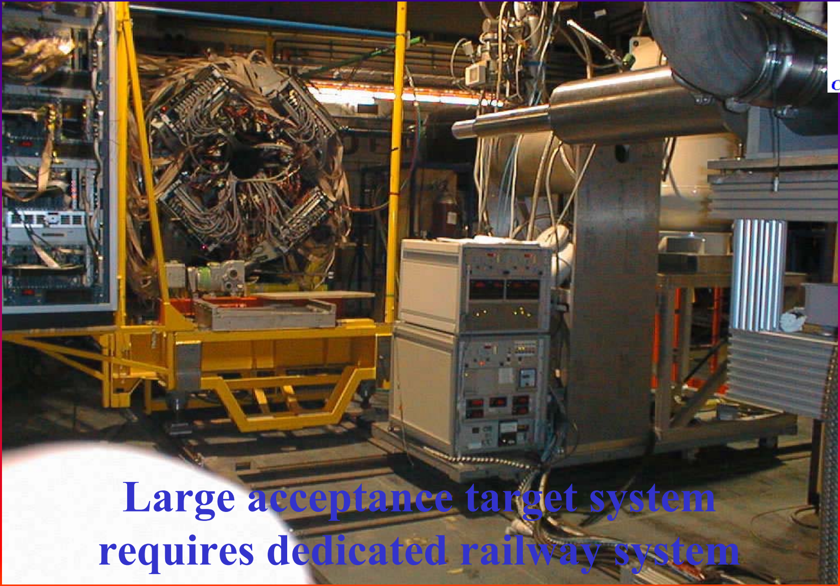
‘frozen spin polarized target’

‘measurement of the GDH sum rule’ (Bonn/Mainz 1998 – 2003)

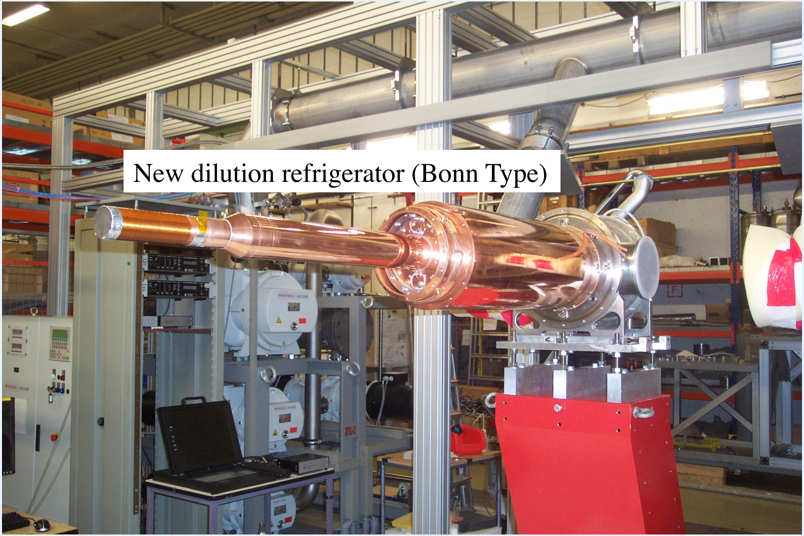
design and set-up : polarized target group‘ PI Bonn
[NIM A 436 (1999) 430]

‘frozen spin polarized target’

‘measurement of the GDH sum rule’ (Bonn/Mainz 1998 – 2003)



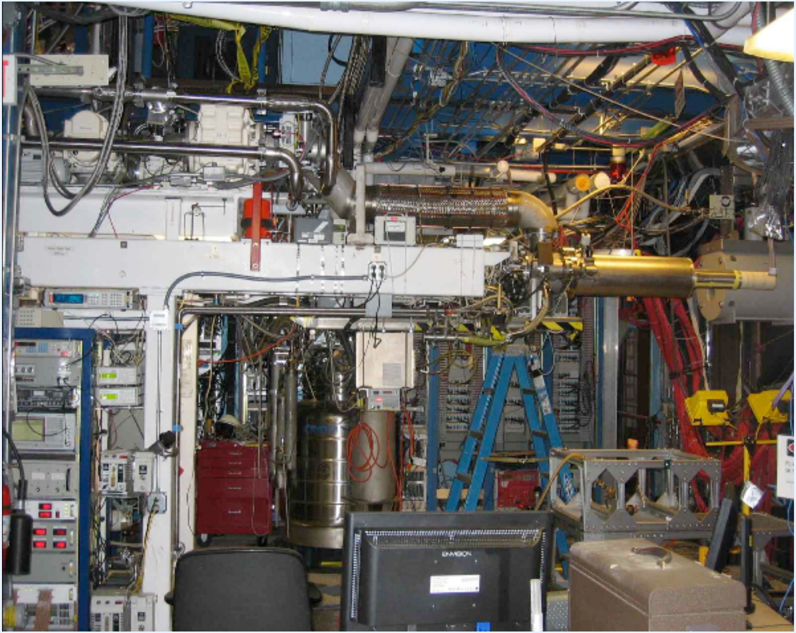
Polarized Target for Crystal Ball at Mainz



Courtesy of A. Thomas

Mainz, Dubna: Frozen Spin Target (20mK and 1 Tesla holding field)
Nucleon Relaxation time: Several 1000h

FROST in Hall B at JLab



Courtesy of Chr. Keith

Frozen Spin Targets for Photoproduction Experiments

Longstanding request:'Complete experiment' e.g. in pion photoproduction $\gamma N \longrightarrow \pi N$

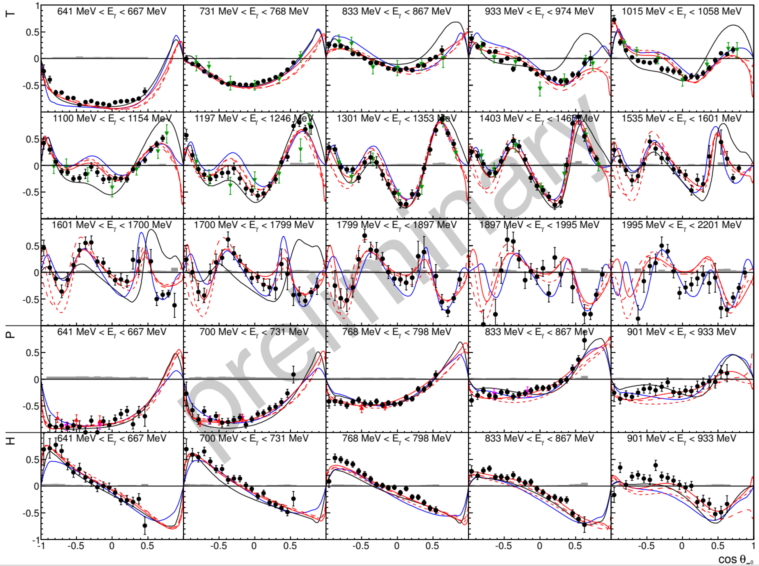
$$\frac{d\sigma}{d\Omega} = \frac{d\sigma_0}{d\Omega} \left\{1 - p_\gamma \Sigma \cos 2\Phi + p_y T + p_\gamma p_z G \sin 2\Phi - p_\gamma p_x H \sin 2\Phi - p_\gamma p_y P \sin 2\Phi\right\}$$

$\frac{d\sigma_0}{d\Omega}$ = unpol. cross sect.
p_γ= γ-pol
Φ=angle between x-z plane and electr. vector of photons
$\vec{p} = \underbrace{(p_x, p_y, p_z)}$ = pol. vector of target nucleons $\longrightarrow \oplus 4\pi$ detection?

↳ in operation at Bonn, Mainz, JLab ↗

Some Polarization Data from Bonn

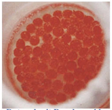
$\gamma p \rightarrow p\pi_0$



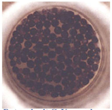
Situation of Target Materials (1)

1979s Proton target materials can be polarized almost completely

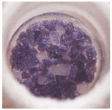
- at every experimental condition
  2.5–5.0T/100mK 5T/1K



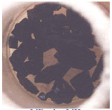
Butanol mit Porphyrexide



Butanol mit CrV complexes



Ammonia



LiD oder LiH

2000s So far this was not the case for deuterated materials

- 'lower' magnetic moment of the deuteron
- use of radicals sufficient to cool protons (EST-concept)

Situation of Target Materials (2)

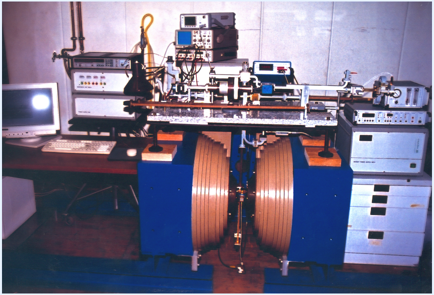
- Borghini Model $\rightarrow$ Equal Spin Temperature (EST) $\rightarrow$ ...width of the electron spin resonance (EPR) line
  $\Longrightarrow$ Radicals opimized for cooling of deuterons
- How do those radicals look like?
  Recipe:
  - Radical production by irradiation, if HFS interaction is weak $\rightarrow$ deuterated materials
  - Chemical doping by use of 'narrow EPR radicals':
    Trityl radicals
    J.H. Ardenkjear-Larsen et al, Proc.Natl.Acad.Sci. USA 100(18)(2003)10158

Additional to the Equipment

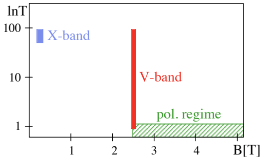
- high magnetic field
- microwave field for DNP
- low temperature cryostat
- NMR for pol. measurements

EPR-Apparatus for paramagnetic radical studies

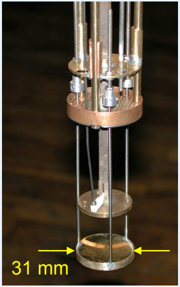
Standard: X-Band Spectrometer at 0.35 T → **9.35 GHz (77 K)**



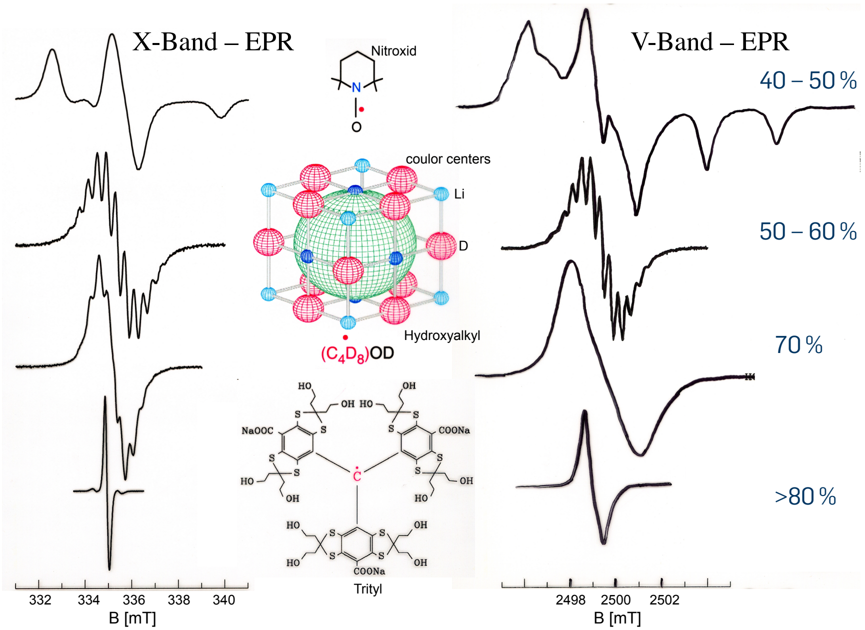
new in Bochum (2005)
V-Band EPR-Spectrometer
➥ 70 GHz at 2.5 T
(300 K → 1 K)



EPR-insert for the ^{4}He-Kryostat with Fabry-Perot resonator

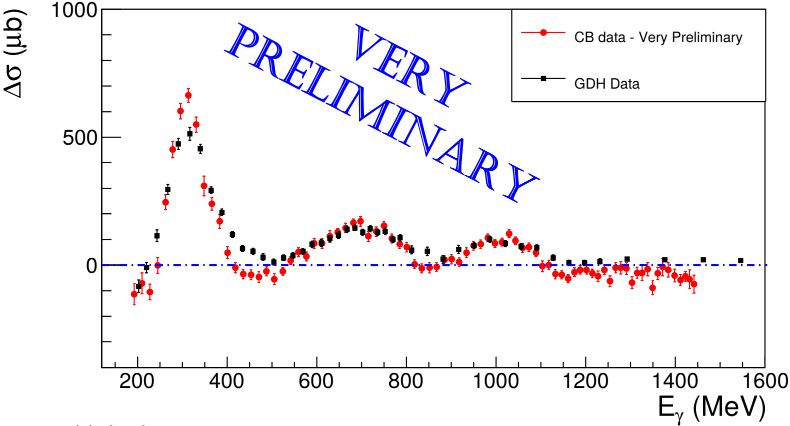


Radicaltyp


X-Band – EPR
V-Band – EPR
Nitroxid
N
O
coulor centers
Li
D
Hydroxyalkyl
$(C_4D_8)OD$
NaOOC
COONa
HO
OH
S
C
Trityl
40 – 50 %
50 – 60 %
70 %
>80 %
332
334
336
338
340
B [mT]
2498
2500
2502
B [mT]

$\sigma_p - \sigma_a \; (\mu b)$

$\vec{\gamma}\,\vec{p} \rightarrow X$



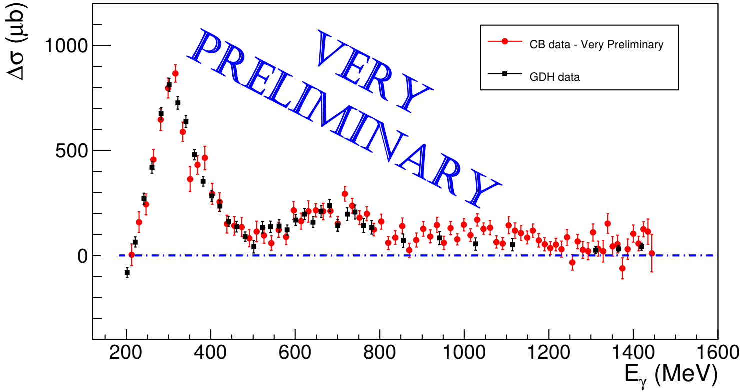
■ Published GDH data - PRL 87, 022003 (01) PRL 91, 192001 (03)

■ A2 Data – VERY PRELIMINARY

(almost the total statistics for dec.2013)

$\sigma_p - \sigma_a\ (\mu b)$

$$\vec{\gamma}\,\vec{d} \rightarrow X$$



■ Published GDH data - PRL 94, 162001 (05) PLB 672, 328 (09)

■ A2 Data – VERY PRELIMINARY

About 30% of the total statistics

Next Challenge for Polarized Solid Targets (1)

- Construction of a '4π continuous mode' polarized target

  ↳ fixed target position inside detector with no moving systems (huge external coils) for the polarization process (DNP)

- In practice: Manufacture of a thin 'internal' superconducting coil with a sufficient field homogeneity for the DNP process
  $\rightarrow$ B $\sim$2.5T; $\Delta B/B < 10^{-4}$
  $\rightarrow$ Thin enough to minimize particle absorption

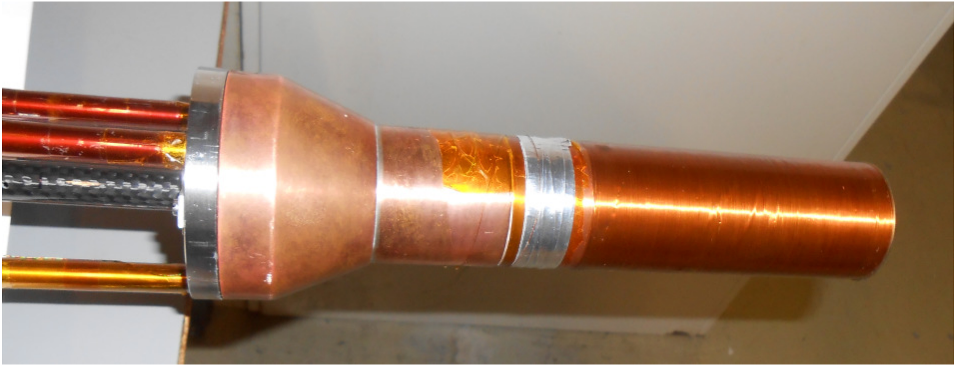
Next Challenge for Polarized Solid Targets (2)

Advantages:

1. No repolarization $\rightarrow$ higher mean polarization
2. No moving systems $\rightarrow$ higher beam time efficiency
3. Higher acceptable beam intensity $\rightarrow N \sim 10^{10} s^{-1} \rightarrow L \sim 10^{33} (\mathrm{cm} \cdot \mathrm{s})^{-1}$

Design of low mass polarizing solenoid
$10^{-4} < \Delta B/B < 10^{-3}$
for the existing Mainz refrigerator

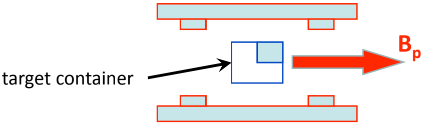
Magnetic field distribution for a corrected coil. (10layer+1 correction layer)



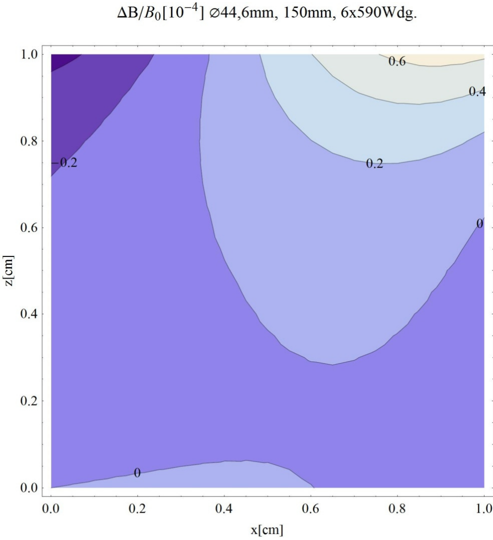
Expected magnetic field at (z=0,r=0) is 2.5T @ 46A and $\Delta B/B \sim 10^{-5}$

Design of low mass polarizing solenoid
$10^{-4} < \Delta B/B < 10^{-3}$
for the new Bonn refrigerator

corrected solenoid : ‘inverse notched coil’,
B = 2.5T @ 90 A



- $\Delta B/B < 10^{-4}$
- 6 layers, 0.25F54 1:1.35, 590 wdg.
- 2 corrector notches (2 * 8 windings)
- Thickness 1.8 mm
- High accuracy winding is mandatory



DNP Solid Targets

From basic research over ~50 years to
an adopted innovative technology

➥ Spin oriented ^{13}C-or ^{15}N-nuclei for medical diagnostics

Important development: Dissolution unit (Malmö 2002) for frozen material

➤ Optimized production path for hyperpolarized ^{13}C-labelled contrast agents

Polarize samples with ^{13}C , ^{6}Li, ^{15}N in solid state (1.2 K /3.5 T)
dissolve rapidly and inject into imager (9.4 T)

conduct *in vivo* MRI experiments with x 10'000 signal improvement

Dissolution - DNP for MRI / NMR

Sdnpi
Swiss dnp Initiative

PAUL SCHERRER INSTITUT PSI

EPFL ÉCOLE POLYTECHNIQUE FÉDÉRALE DE LAUSANNE

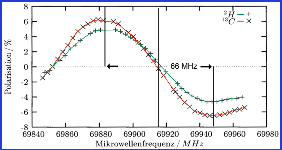
Latest main activities: San Francisco (USA) – first applications at human beings – and Lausanne (Switzerland)

Bochum / GE Healthcare

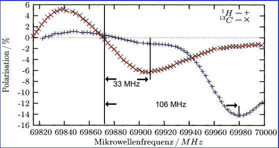
Focus:

1) deut. pyruvic acid: C_3D_3OH + Triphenylmethyl radical (Trityl radical) AH 11501
   → DNP-Mechanism (Solid State Effect - EST)

frequency curves for



deuterons and ^{13}C

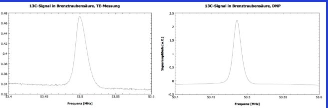


protons and ^{13}C

2) Polarization enhancement of ^{13}C in C_3H_3OH + AH 11501

3.35 T / 1.2 K → 5.0 T / 1.0 K

$P_{^{13}C}$: 35% → 75%



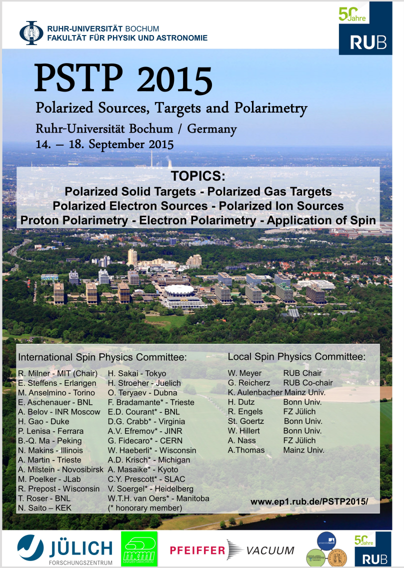
$P_{^{13}C}$ (T.E.) = 0.13% $P_{^{13}C}$ (DNP) = 75%

→ in vivo MRI → enhancement factor: 50000

Bright future for
Polarized Solid Targets

(after very successful operation in this field of particle physics since ~ 50 years, nowadays in the field of medicine, too)

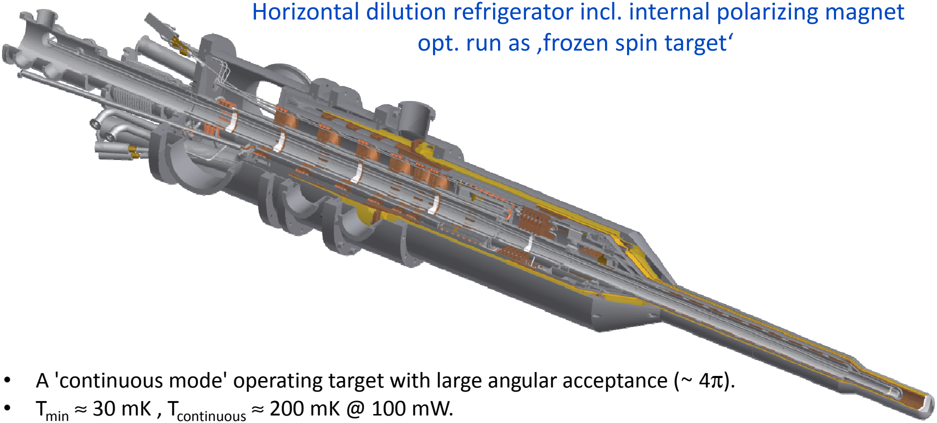
Welcome to Bochum!!!



Appendix

Low-T refrigerator with high current leads
Bochum/Bonn

Horizontal dilution refrigerator incl. internal polarizing magnet
opt. run as ‚frozen spin target‘



- A 'continuous mode' operating target with large angular acceptance (~ 4π).
- $T_{min} \approx 30$ mK , $T_{continuous} \approx 200$ mK @ 100 mW.
- High luminosity L ~ 10^{33}/cm²s (N ≈ 10^{10}/s).
- High mean polarization.
- Equipped with an internal superconducting polarizing magnet for permanent DNP and high current leads.

Why internal polarizing coil?

- Target re-polarization; loss of beam time & problem in reproducing detector position
- Replace the external magnet- polarize the target during experiment
- It must fulfill the following conditions;
  - ✓ High homogeneity $\leq 10^{-4}$
  - ✓ Magnetic field ≈ 2.5 T
  - ✓ Thin enough to minimize particle absorption

Theoretically this is achievable by;

- Ten layer coil with two layer correction & a current of 46 A

The Principle of Dynamic Nuclear Polarization

Basic principle:

State of lowest energy is favoured

$$P = \frac{\langle I_z \rangle}{I_z^{max}} = B_I\left(\frac{\mu B}{2kT}\right) \overset{2.5T,\ 1K}{=} 0.25\% \text{ (Proton)}$$

$$= 0.05\% \text{ (Deuteron)}$$

⇒ ‚Brute Force' : Maximize B and Minimize T

Trick:

Transfer of polarization from particles with high μ

Electrons: $P \overset{2.5T,\ 1K}{=} 93\%$

Doping with paramagnetic centers:

~ 10^3 nuclei feeded by 1 unpaired electron from:

- ♦ Chemically stable radical → Liquids
- ♦ Radiation induced defects → Solids

Summary of DNP

- **Proton target materials can be polarized almost completely**
  – under almost every condition
  2.5 - 5.0 T / 100 mK 5 T / 1 K
  – independently of the actual material
  H-butanol, H-propanediol, NH_3

- **So far this was not the case for the deuterated materials**
  (as we have seen)
  – 'Low' magnetic moment of the deuteron
  – Use of radicals sufficient to cool protons

⇨ **Radicals optimized for cooling of deuterons**

How do those Radicals look like ? (1)

❖ **Theory**: Optimize the strength of their non-Zeeman interactions

How? Minimize the homogeneous dipolar width D of the Electron-Zeeman state

$$D \sim B_\ell \approx g_e \mu_B \cdot N_S \qquad N_S = \text{number of Spins}$$

Is this enough ? — No !

In practice:

Inhomogeneous interactions present
– Anisotropy of the g-factor (magn. field dependent)
– Hyperfine interaction (magn. field independent)

$$D^2 \sim B^2_{\ell,\text{hom.}} + B^2_{\ell,\text{inhom.}}$$

↳ exp. determination by EPR-measurements

Michel Borghini

some personal reflections

EXPERIENCE WITH NH_3 AS TARGET MATERIAL FOR POLARIZED PROTON TARGETS AT THE BONN 2.5 GEV ELECTRON SYNCHROTRON

U. Härtel, O. Kaul, W. Meyer, K. Rennings, E. Schilling
Physikalisches Institut der Universität Bonn, Germany

Conclusion

We have found a reproducable method to create paramagnetic radicals in solid ammonia by irradiation in a 20 MeV electron beam, cooling the samples in liquid argon. No disintegration of the beads into powder has been observed so far. The so prepared ammonia could be polarized with a relatively short polarization build-up time. Taking also into account the good radiation resistance and the possibility of annealing ammonia seems to be a useful target material for high energy physics experiments.

Michel Borghini

some personal reflections

FIRST DYNAMIC DEUTERON POLARIZATION MEASUREMENTS IN IRRADIATED ND_3

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ABSTRACT

Dynamic deuteron polarization has been demonstrated in deuterated ammonia, using paramagnetic radicals created by electron irradiation. During the irradiation, performed in the 20 MeV electron beam of the Bonn injection linac, the solid ammonia beads were cooled in liquid argon at a temperature of 87 K. A deuteron polarization of 11% was obtained in a field of 2.5 T and at a temperature of 0.5 K. These first measurements with an irradiated ND_3 sample indicated a rather small electron spin resonance line. The deuteron tensor polarization could be changed by a simple method.

It seems, that in irradiated ND_3 samples several polarization mechanisms take place. Further investigations are needed to understand all phenomena completely.

‘continuous mode polarized target’

Continuous cooling : ^{4}He evaporation cryostat (SACLAY 1966)

P. Roubeau Cryogenics 6 (1966) 207

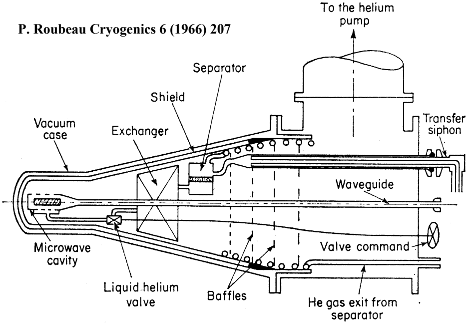


Figure 3. Schematic diagram of the cryostat

TECHNOLOGY OF HIGH ENERGY TARGETS

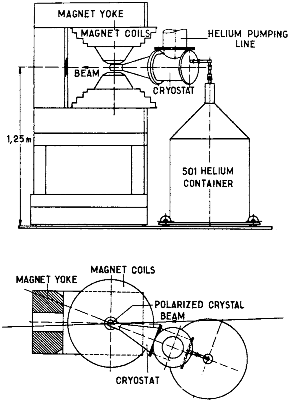


Fig. 14 CERN polarized target [16].

1st continuous cooling ^{4}He cryostat

‘Roubeau – cryostat’

T_{min} ~ 1K, Q ~ 400mW

P_{max} ~ 40 % @ 2.5 T

Target material : i.e. butanol

CERN polarized target

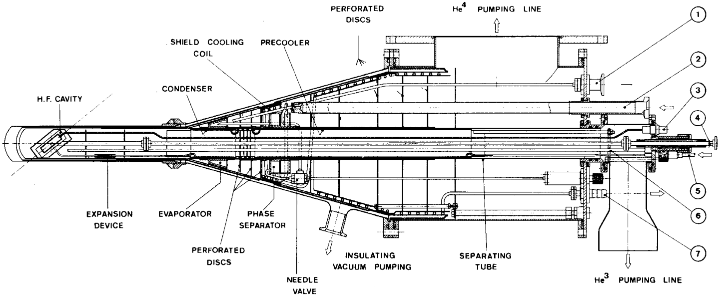
Borghini et al. (1969)

‘continuous mode polarized target’

Continuous cooling : ^{3}He – evaporation cryostat (1970)

48 P. ROUBEAU

P. Roubeau, IInd Int. Conf. on Pol. Targets, LBL (1971) 47



Roubeau‘s ^{3}He – cryostat (CERN 1970):

T_{min} ~ 0.4 K, Q ~ 50 mW @ 0.655 K with 250 m³/h p.s.

P_{max} ~ 65 % - ~ 90 %

‘frozen spin polarized target’

‘limitations of the frozen spin principle

‘Frozen Spin Target’ :

- good angular acceptance (~ 4π)
- moderate luminosity L ~ $10^{30}/cm^2s$ (N ≈ 10^7/s)
- moderate mean polarization
- moderate beam time efficiency

‘continuous mode’ target :

- bad angular acceptance (#msrad)
- high luminosity L ~ $10^{35}/cm^2s$ (N ≈ 10^{12}/s)
- high mean polarization
- good beam time efficiency

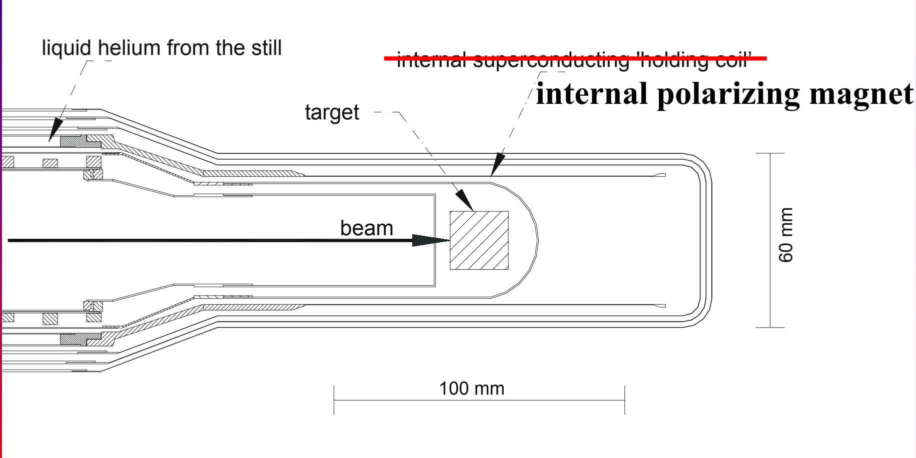
Scope : combine both concepts

‘4π - continuous mode’ target :

- good angular acceptance (~ 4π)
- high luminosity L ~ $10^{33}/cm^2s$ (N ≈ 10^{10}/s)
- high mean polarization
- Good beam time efficiency

New concepts

‘4π continuous mode target’

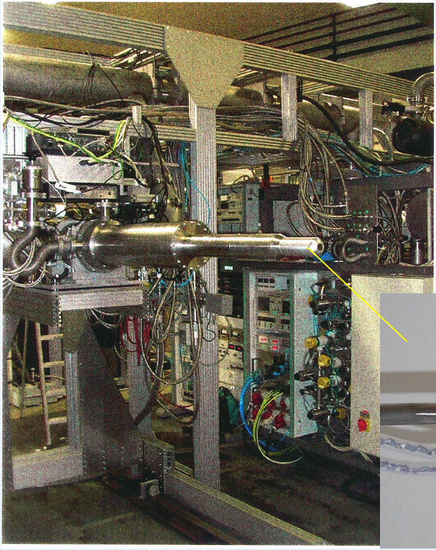


$\varnothing$ 44 mm, l ~ 160 mm, d ≤ 1.5 mm

goal : B_p ~ 2.5 Tesla, $\Delta B/B \sim 10^{-4}$

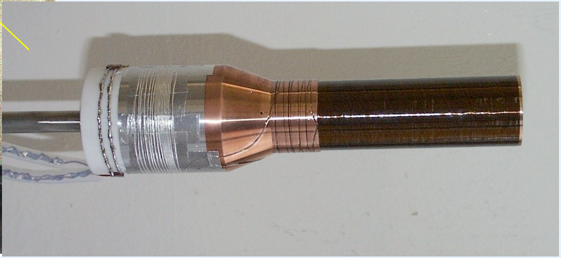
GDH-Experiments at Mainz, Bonn

Marriage of polarized solid target (Frozen SPIN TARGET) and 4π-particle detection decisive (Bonn 1998)



GDH sum rule at the proton ✓
GDH sum rule at the neutron (< 1.8 GeV performed)
↑
80% in D-butanol + Trityl radical (Bochum 03)

0.64T magnetic ‚holding‘ coil



Future Developments

Longstanding request: 'Complete experiment' e.g. in pion photoproduction $\gamma N \rightarrow \pi N$

$$\frac{d\sigma}{d\Omega} = \frac{d\sigma_0}{d\Omega} \left\{ 1 - p_\gamma \Sigma \cos 2\phi + p_y T + p_\gamma p_z G \sin 2\phi - p_\gamma p_x H \sin 2\phi - p_\gamma p_y P \sin 2\phi \right\}$$

$\frac{d\sigma_0}{d\Omega}$ = unpol. cross sect.; p_γ = γ-pol; ϕ = angle between x-z plane and electr. vector of photons

$\vec{p} = (p_x, p_y, p_z)$ = pol. vector of target nucleons → ⊕ 4π detection?

(p_x, p_y): see next

p_z: in operation at Bonn, Mainz, JLab